

Canonical structure of dynamical fluctuations in mesoscopic nonequilibrium steady states

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We give the explicit structure of the functional governing the dynamical density and current fluctuations for a mesoscopic system in a nonequilibrium steady state. Its canonical form determines a generalised Onsager-Machlup theory. We assume that the system is described as a Markov jump process satisfying a local detailed balance condition such as typical for stochastic lattice gases and for chemical networks. We identify the entropy current and the traffic between the mesoscopic states as extra terms in the fluctuation functional with respect to the equilibrium dynamics. The density and current fluctuations are coupled in general, except close to equilibrium where their decoupling explains the validity of entropy production principles.

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Fluctuation theory is at the heart of statistical mechanics. About a century ago appeared the first fluctuation formulae regarding equilibrium systems, in particular from Boltzmann's statistical interpretation of the thermodynamic entropy. As example, the equilibrium density fluctuations of a gas in large volume V at inverse temperature β and chemical potential μ satisfy the asymptotic law

$$\mathbf{P}\left(\frac{N}{V} \simeq n\right) \sim e^{-\beta V [\Omega(\mu, n) - \Omega(\mu)]} \quad (1)$$

where $\Omega(\mu)$ is the grand potential and $\Omega(\mu, n) = F(n) - \mu n$ with $F(n)$ the free energy, is the corresponding variational functional; at least away from the phase coexistence regime where droplet formation or nucleation mechanisms become responsible for a slower, surface-exponential decay. Yet in all cases, there appears an important relation between the structure of equilibrium fluctuations and the thermodynamics of the system, making the equilibrium domain exceptionally well understood. In particular, the variational principles characterising equilibrium can be understood as an immediate consequence of its fluctuation theory and response relations can be derived from expanding (1) around the equilibrium density n_0 .

In order to include dynamics in the fluctuation theory, Onsager and Machlup derived the generic structure of small time-dependent equilibrium fluctuations and explained how their dynamics relates to the return to equilibrium, [1]. The ensuing linear response theory formalised the general relation between equilibrium current fluctuations and the response in driven systems in a first-order perturbation theory around equilibrium. To go beyond and challenged by *e.g.* the fast progress in nonequilibrium experiments on nanoscale, one soon realises a

lack of general principles. Moreover it would be too optimistic to think nonequilibrium physics based solely on quantities typical to equilibrium descriptions supplemented with the corresponding currents. Deeply related to that is the lack of generally valid variational principles for nonequilibrium steady states, beyond the approximate ones of minimum/maximum entropy production. Yet, more recently there has also been great progress. One well-known approach to dynamical (and especially current) fluctuations in open systems adds to the models fields representing the various reservoirs that count the long-time statistics of associated 'charges' by Master equation or stochastic path methods, see *e.g.* [2, 3, 4]. The hydrodynamic fluctuations for some stochastic lattice gas models have been studied in *e.g.* [5, 6]. For some standard lattice gas models the large deviations can in fact be explicitly calculated, see the review [7]. Up to now, special emphasis was put on the fluctuations of the current, also because of relations with a celebrated fluctuation symmetry of the entropy production, [8, 9].

In the present letter we come back to the basic question whether there is at all any systematics in the fluctuations beyond equilibrium or close-to-equilibrium. Can one develop a formalism that would—similarly to the equilibrium scheme—establish a link between the dynamical fluctuations and mean (thermo-)dynamical properties of a system, possibly with the entropy production playing a role similar to the entropy at equilibrium? And could that also explain the appearance and limitations of the entropy-production variational principles on a fluctuation basis? As we have shown before, [10], the minimum entropy production principle close to equilibrium follows from the fluctuation theory for the occupation (or residence) times, which are the relative times spent at different states of the system. This supports both the relevance of dynamical fluctuation theory for understanding the status and the validity of various nonequilibrium variational principles, and the importance of time-symmetric observables in these considerations. Indeed our results here strongly suggest that only by treating

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jointly the time-symmetric and -antisymmetric sectors do model-specific results make place for a unique fluctuation structure. As far as we know, that is one of the rare occasions where the nonequilibrium world can be seen submitted to general laws.

To address the above questions and in the context of a stochastic network we propose to study the joint dynamical fluctuations of the occupation times (time-symmetric) and currents (time-antisymmetric). We will show that these joint fluctuations have an explicit and general structure, with a fluctuation functional derived from the so-called traffic measuring the mean dynamical activity in the system, which is hence the counterpart of the entropy or grand potential in the equilibrium static fluctuation theory, *cf.* (1). Only close to equilibrium there emerges a simple relation between that traffic and the entropy production. Together with a decoupling between small time-symmetric and time-antisymmetric fluctuations in the close-to-equilibrium domain, this lies behind the approximate validity of the entropy production principles. This substantially extends the argument in [10]. Our main results are relations (11), (17), and (27) below. The application of our formalism but for driven diffusions can be found in [11]. Our emphasis here on jump processes makes the analysis also suitable to the statistics of quantum transport as *e.g.* in [3, 12]; particular examples will follow separately.

In contrast with full counting statistics methods, the reservoirs are not made explicit in our approach. Instead, we assume that the changes in all reservoirs or loads are mutually distinguishable and can be read off from the trajectory of the system. Another remark concerns the meaning of the occupation times. This is a dynamical observable and its fluctuations fundamentally differ from static fluctuations, which count the plausibility that the system obeys some statistics given that the system was in its typical stationary state far in the past. These static fluctuation functionals are nonequilibrium variants of the equilibrium free energy and have been extensively studied in the context of lattice gases in the hydrodynamic limit, [13]. As a matter of principle, one expects that one could recover the static from the dynamical fluctuations that are studied here.

The mathematics involved is the theory of large deviations and stems from the work of Donsker and Varadhan, [14]. A useful and repeatedly exploited technique in this approach is to compute the fluctuation functionals on more coarse-grained levels from constrained minimisations of a fine-grained functional. That is called the contraction principle.

I. GENERAL FORMALISM

We consider a mesoscopic nonequilibrium system modelled by a stationary ergodic process X_t making jumps on a discrete set of states, $\{x, y, \dots\}$. As is typical for a thermodynamic formalism it is not essential for the math-

ematical structure whether the process represents a single particle random walk or a many-body open system. We only ask the dynamics as given by some transition rates $w(x, y)$ on ordered pairs $x \rightarrow y$ to be ergodic. For an easy interpretation we assume that the local detailed balance principle applies, according to which $\log [w(x, y)/w(y, x)]$ is the entropy change in the environment (possibly made of several distinct reservoirs) per single event $x \rightarrow y$. Tracing the whole trajectory of the system, all currents as well as the total entropy exchange with the environment can be determined.

We start from that fine-grained level of description and we consider as dynamical observables the occupation times

$$p_T(x) = \frac{1}{T} \int_0^T \chi(\omega_t = x) dt \quad (2)$$

(with χ equal to one or zero, indicating whether the event in brackets occurs, respectively does not occur) jointly with the two-point correlations, for all $x \neq y$,

$$C_T(x, y) \delta t = \frac{1}{T} \int_0^T \chi(\omega_t = x) \chi(\omega_{t+\delta t} = y) dt \quad (3)$$

counting the number of jumps $x \rightarrow y$, both defined for each realisation of the process $(\omega_t; 0 \leq t \leq T)$. The occupation times $p_T(x)$ form a random distribution that asymptotically approaches the stationary distribution, $\lim_{T \rightarrow \infty} p_T(x) = \rho(x)$, with probability one by the ergodic theorem. Similarly, the empirical correlations have the almost-sure asymptotics $\lim_{T \rightarrow \infty} C_T(x, y) = \rho(x)w(x, y)$.

The question about dynamical fluctuations concerns the long-time asymptotics of possible deviations of p_T and C_T from their typical values: to compute the probability $\mathbf{P}_T(p, k)$ to observe for all x and y ,

$$p_T(x) = p(x), \quad C_T(x, y) = p(x)k(x, y) \quad (4)$$

We must add here the stationarity condition $\sum_y [p(y)k(y, x) - p(x)k(x, y)] = 0$ since, by conservation of probability, $\lim_{T \rightarrow \infty} \sum_y [C_T(x, y) - C_T(y, x)] = 0$ for every realisation of the process. To determine $\mathbf{P}_T(p, k)$ of (4), we compare the path-distribution of the original stationary process with rates w to a fictitious stationary process with rates k . The former distribution reads, with $\lambda(x) = \sum_y w(x, y)$ the escape rates,

$$\begin{aligned} \mathbf{P}_T(\omega) = \rho(x_0) e^{-\lambda(x_0)t_1} w(x_0, x_1) dt_1 e^{-\lambda(x_1)(t_2-t_1)} \dots \\ \dots w(x_{n-1}, x_n) dt_n e^{-\lambda(x_n)(T-t_n)} \end{aligned} \quad (5)$$

on realisations $\omega = (x_0, 0; x_1, t_1; \dots; x_n, t_n \leq T)$ with jumps $x_{k-1} \rightarrow x_k$ at times t_k . The probability $\mathbf{P}_T^*(\omega)$ of the same realisation under the fictitious process is obtained by replacing w with k , and λ with the escape rates $\sum_y k(x, y)$. We exploit that (i) for any trajectory ω satisfying the constraints (4) the density of \mathbf{P}_T with respect

to the \mathbf{P}_T^* only depends on the time-averages $p(x)$ and $k(x, y)$; (ii) those values p and k become *typical* under the fictitious process \mathbf{P}_T^* when $T \rightarrow \infty$. Using both properties, the probability under study is

$$\begin{aligned} \mathbf{P}_T(p, k) &= \mathbf{P}_T^*(p, k) \left\langle \frac{d\mathbf{P}_T}{d\mathbf{P}_T^*} \mid \text{conditions (4)} \right\rangle_{\mathbf{P}_T^*} \\ &\sim \frac{d\mathbf{P}_T}{d\mathbf{P}_T^*}(p, k) \end{aligned} \quad (6)$$

asymptotically for $T \rightarrow \infty$. Explicitly, $\mathbf{P}_T(p, k) \sim \exp[-T\mathcal{I}(p, k)]$ with the fluctuation functional

$$\mathcal{I}(p, k) = \sum_{x,y} p(x) \left[k(x, y) \log \frac{k(x, y)}{w(x, y)} - k(x, y) + w(x, y) \right] \quad (7)$$

(remember that $I(p, k) = \infty$ whenever p is not stationary with respect to the transition rates k). This result is our starting point towards a systematic generation of various other fluctuation laws by contraction, in both the time-symmetric and the time-antisymmetric domains.

II. OCCUPATION-CURRENT FLUCTUATIONS

The observed time-averaged currents correspond to the antisymmetric part of the two-point correlations, $C_T(x, y) - C_T(y, x)$. The joint fluctuation law for the currents and the occupation times

$$\mathbf{P}_T(p, j) \sim e^{-T I(p, j)} \quad (8)$$

can be derived from (7) by solving the minimisation problem

$$I(p, j) = \inf_k \{ \mathcal{I}(p, k) \mid p(x)k(x, y) - p(y)k(y, x) = j(x, y) \} \quad (9)$$

For stationary currents, $\sum_y j(x, y) = 0$, to which we from now on solely restrict, the solution $I(p, j) = \mathcal{I}(p, k^*)$ is determined from

$$\begin{aligned} k^*(x, y) &= w(x, y) e^{\Delta(x, y)/2} \\ \Delta(x, y) &= -\Delta(y, x) \\ j(x, y) &= p(x)k^*(x, y) - p(y)k^*(y, x) \end{aligned} \quad (10)$$

(Otherwise $I(p, j) = \infty$.) As a result,

$$I(p, j) = \frac{1}{4} \sum_{x,y} \Delta(x, y)j(x, y) - \frac{1}{2} \sum_{x,y} [t_p^*(x, y) - t_p(x, y)] \quad (11)$$

in which

$$t_p(x, y) = p(x)w(x, y) + p(y)w(y, x) \quad (12)$$

and

$$t_p^*(x, y) = p(x)k^*(x, y) + p(y)k^*(y, x) \quad (13)$$

measure the mean dynamical activities; we call them *traffic* and they yield the symmetric counterpart to the expected currents. The second term in (11) is therefore an *excess* in the overall traffic needed to create the fluctuation or to make it typical. Similarly, by the local detailed balance principle, the first term corresponds to an excess in the entropy flow to the environment which amounts to $\dot{S} = \frac{1}{2} \sum_{x,y} j(x, y) \log[w(x, y)/w(y, x)]$ under the original process and analogously for the modified one.

Next, being motivated by the equilibrium fluctuation theory, *cf.* (1), we reveal a hidden canonical structure that enables a particularly illuminating formulation of our result. Any nonequilibrium process can be related to a reference detailed balanced one with rates $w_0(x, y)$, so that $w(x, y) = w_0(x, y) e^{\sigma(x, y)/2}$ with some driving $\sigma(x, y) = -\sigma(y, x)$. (For example, the rates $w_0(x, y) = \sqrt{w(x, y)w(y, x)} e^{s(y)-s(x)}$ and s an arbitrary state function, can serve as such a reference.) Having fixed w_0 , the rates $w(x, y) = w_\sigma(x, y)$ are now parameterised by the driving $\sigma(x, y)$, and we introduce the potential function

$$H(p, \sigma) = 2 \sum_{x,y} p(x) [w_\sigma(x, y) - w_0(x, y)] \quad (14)$$

equal to the excess in the overall traffic with respect to that reference. It is a potential for the expected transient currents $j_{p,\sigma}(x, y) = p(x)w_\sigma(x, y) - p(y)w_\sigma(y, x)$ in the sense that

$$\delta H(p, \sigma) = \frac{1}{2} \sum_{x,y} j_{p,\sigma}(x, y) \delta\sigma(x, y) \quad (15)$$

(with the p kept fixed in the variation). Its Legendre transform is

$$G(p, j) = \sup_{\sigma'} \left[\frac{1}{2} \sum_{x,y} \sigma'(x, y)j(x, y) - H(p, \sigma') \right] \quad (16)$$

and we observe that the supremum (taken over all antisymmetric matrices) is attained at $\sigma' = \sigma^*$ such that $j_{p,\sigma^*} = j$, which means that the driving σ and the current j are canonically conjugated variables. Further, eqs. (10) are solved with $\Delta = \sigma^* - \sigma$, hence the fluctuation functional $I(p, j) = I_\sigma(p, j)$, eq. (11), obtains the final form

$$I_\sigma(p, j) = \frac{1}{2} [G(p, j) + H(p, \sigma) - \dot{S}(\sigma, j)] \quad (17)$$

with

$$\dot{S}(\sigma, j) = \frac{1}{2} \sum_{x,y} \sigma(x, y)j(x, y) \quad (18)$$

the observed entropy current into the environment. That is our main result, giving the fluctuation functional entirely in terms of the potential function $H(p, \sigma)$ (*i.e.* in terms of the overall traffic) and derived quantities. The functional $G(p, j)$ directly gives the reference equilibrium dynamical fluctuations as $I_0(p, j) = \frac{1}{2}G(p, j)$, hence (17)

specifies the nonequilibrium correction to that equilibrium. Remark also that the antisymmetric part of the functional I_σ under time reversal equals $I_\sigma(p, -j) - I_\sigma(p, j) = \dot{S}(\sigma, j)$, compare [9, 15, 16], which is just the steady state fluctuation symmetry. However, more important is that (17) also in a generic way specifies the time-symmetric component. That is why (17) represents a generalised Onsager-Machlup Lagrangian describing steady fluctuations, the generalised dissipation functions being G and H . At the same time, one recognises the mathematical structure of equilibrium fluctuations; the grand potential $\Omega(\mu)$ and the variational functional $\Omega(\mu, n)$ of (1) get replaced here by $-H(p, \sigma)/2$ and $[G(p, j) - \dot{S}(\sigma, j)]/2$, respectively. Note that while the potentials G and H depend on the choice of reference equilibrium dynamics, the resulting functional $I(p, j)$ is of course independent of that.

Fluctuation laws on a more-coarse grained level, *e.g.*, the fluctuations of a single selected current, can be obtained by further contractions starting from (7) or (17). Then, depending on the particular question, a modified canonical formalism can be established.

There is a trivial yet important generalisation of the above results to systems in which a transition $x \rightarrow y$ can go via multiple channels, each possibly corresponding to the interaction with different reservoirs. For these systems the formulas (7), (11), (15), (16) etc remain valid if the ordered pairs x, y in the sums get replaced with x, y, α , the α labelling the channels. A simple example of such a multi-channel model comes in the next section.

III. EXAMPLE

We demonstrate the above formalism on a model of transport over a single level (quantum dot). There are two configurations $x = 0, 1$ corresponding to the level being empty respectively occupied, and it is coupled to the left (L) and the right (R) reservoirs. Using the notation V_L and V_R for the potential gradients between that level and the reservoirs, both oriented in the $L \rightarrow R$ direction, the local detailed balance principle restricts the possible transition rates corresponding to each channel to the following general form:

$$\begin{aligned} w_L(0, 1) &= \Gamma_L e^{\beta V_L/2}, & w_L(1, 0) &= \Gamma_L e^{-\beta V_L/2} \\ w_R(0, 1) &= \Gamma_R e^{-\beta V_R/2}, & w_R(1, 0) &= \Gamma_R e^{\beta V_R/2} \end{aligned} \quad (19)$$

For simplicity, we consider here only the case $\Gamma_L = \Gamma_R = \Gamma$. Writing the occupation times as $p(0) = (1 - v)/2$ and $p(1) = (1 + v)/2$, the expected transient currents (also both oriented in the $L \rightarrow R$ direction) and traffic separately for each channel equal

$$j_v^{L,R} = \Gamma \sinh \frac{\beta V_{L,R}}{2} \mp \Gamma v \cosh \frac{\beta V_{L,R}}{2} \quad (20)$$

$$t_v^{L,R} = \Gamma \cosh \frac{\beta V_{L,R}}{2} \mp \Gamma v \sinh \frac{\beta V_{L,R}}{2} \quad (21)$$

As a reference equilibrium we take the dynamics (19) for $V_L = V_R = 0$ (with the symmetric part Γ kept unchanged). The current potential (14) is determined from the overall traffic:

$$H(v, V_L, V_R) = 2\Gamma \left(\cosh \frac{\beta V_L}{2} - v \sinh \frac{\beta V_L}{2} + \cosh \frac{\beta V_R}{2} + v \sinh \frac{\beta V_R}{2} - 2 \right) \quad (22)$$

One checks that $\partial H / \partial V_{L,R} = \beta j_v^{L,R}$ which is an instance of (15). The Legendre transform of H at $j^L = j^R = j$ gives the occupation-current fluctuation functional $G(v, j) = I_0(v, j)/2$ for the reference equilibrium dynamics, *cf.* (16):

$$\begin{aligned} G(v, j) &= \sup_{V_L, V_R} [\beta j(V_L + V_R) - H(v, V_L, V_R)] \\ &= 4j \log \left[\frac{1}{\sqrt{1-v^2}} \left(\frac{j}{\Gamma} + \sqrt{1-v^2 + \frac{j^2}{\Gamma^2}} \right) \right] \\ &\quad + 4\Gamma \left[1 - \sqrt{1-v^2 + \frac{j^2}{\Gamma^2}} \right] \end{aligned} \quad (23)$$

This extends to the nonequilibrium dynamics by the generalised Onsager-Machlup formula (17). *E.g.*, in the L-R symmetric case $V_L = V_R = V$, the entropy flow is $\dot{S} = 2\beta V j$, and the nonequilibrium fluctuation functional becomes

$$\begin{aligned} I_V(v, j) &= 2j \log \left[\frac{1}{\sqrt{1-v^2}} \left(\frac{j}{\Gamma} + \sqrt{1-v^2 + \frac{j^2}{\Gamma^2}} \right) \right] - \beta V j \\ &\quad + 2\Gamma \left[\cosh \frac{\beta V}{2} - \sqrt{1-v^2 + \frac{j^2}{\Gamma^2}} \right] \end{aligned} \quad (24)$$

Due to the ‘particle-hole’ symmetry $I(-v, j) = I(v, j)$, the (marginal) current fluctuations correspond to the rate $\mathfrak{I}_V(j) = I_V(0, j)$, which is

$$\begin{aligned} \mathfrak{I}_V(j) &= 2j \log \left[\frac{j}{\Gamma} + \sqrt{1 + \frac{j^2}{\Gamma^2}} \right] - \beta V j \\ &\quad + 2\Gamma \left[\cosh \frac{\beta V}{2} - \sqrt{1 + \frac{j^2}{\Gamma^2}} \right] \end{aligned} \quad (25)$$

Again by contraction, the fluctuation functional for the occupation times is $\mathfrak{I}_V(v) = I_V(v, j^*)$ where $j^* = \Gamma \sqrt{1-v^2} \sinh(\beta V/2)$ is the most probable value of the stationary current given v . As a result,

$$\mathfrak{I}_V(v) = 2\Gamma \cosh \left(\frac{\beta V}{2} \right) (1 - \sqrt{1-v^2}) \quad (26)$$

IV. REGIME OF SMALL FLUCTUATIONS

The main features of the joint occupation-current fluctuations already become manifest in the leading order around the nonequilibrium steady state. For our original dynamics with stationary distribution ρ , steady current \bar{j} and steady traffic \bar{t} , we write $p = \rho(1 + \epsilon u_1)$,

$j = \bar{j} + \epsilon j_1$. Standard perturbation theory applied to (11), up to quadratic order in ϵ , gives as a final result $I(p, j) = \epsilon^2 I^{(2)}(u_1, j_1)$ where

$$\begin{aligned} I^{(2)}(u_1, j_1) = & \frac{1}{4} \sum_{x,y} \left[\frac{1}{2\bar{t}} j_1^2 + \frac{\bar{t}}{2} (\nabla^- u_1)^2 \right. \\ & \left. - \frac{\bar{j}}{\bar{t}} j_1 \nabla^+ u_1 + \frac{\bar{j}^2}{2\bar{t}} (\nabla^+ u_1)^2 \right] (x, y) \end{aligned} \quad (27)$$

with the shorthand $\nabla^\pm u_1(x, y) = [u_1(x) \pm u_1(y)]/2$. This formula clearly demonstrates how the occupation times and current become coupled away from equilibrium. That coupling is proportional to the stationary current and vanishes only in the close-to-equilibrium regime where $\bar{j} = O(\epsilon)$.

The appearance/disappearance of the occupation-current correlation is deeply related with the validity/breaking of the entropy production principles. The expected value of the (transient) entropy production rate $\mathcal{E}(p)$ at a given distribution p is the sum of the expected entropy flow $\frac{1}{2} \sum_{x,y} j_p(x, y) \log [w(x, y)/w(y, x)]$ and the rate of increase of the system's entropy $-\frac{1}{2} \sum_{x,y} j_p(x, y) \log p(x)$, [15]. In the same quadratic approximation as above but now close to equilibrium so that $w = w_0[1 + O(\epsilon)]$, the entropy production rate equals

$$\mathcal{E}(p) = \sum_{x,y} \left[\frac{\epsilon^2 \bar{t}}{2} (\nabla^- u_1)^2 + \frac{\bar{j}^2}{2\bar{t}} \right] (x, y) \quad (28)$$

with $\bar{j} = O(\epsilon)$. On the other hand, from (27) the marginal distribution of the occupation times for $\bar{j} = O(\epsilon)$ corresponds to the functional $\mathfrak{J}^{(2)}(u_1) = \frac{1}{8} \sum_{x,y} \bar{t} (\nabla^- u_1)^2(x, y)$, and hence

$$\mathfrak{J}(p) = \frac{1}{4} [\mathcal{E}(p) - \mathcal{E}(\rho)] \quad (29)$$

see [10] for more details. Hence, the stationary distribution ρ is a minimiser of the entropy production rate and the latter governs the occupation fluctuations—this is no longer true beyond the close-to-equilibrium regime where the occupation-current correlation becomes relevant. A similar argument reveals a direct link between the current fluctuations and the maximum entropy production principle, [11].

V. CONCLUSIONS AND REMARKS

We have derived an explicit formula for the functional governing the joint dynamical fluctuations of transi-

sition intensities and occupation times in a steady state regime described by a Markov jump process, (7). In the occupation-current form (17), it gets a remarkable canonical structure: the (reference) equilibrium functional is corrected by its Legendre transform which is just a potential for the expected currents, and by the entropy flow. These functionals form a natural starting point towards the study of fluctuations for any selected collection of observables that can be expressed in terms of transitions/currents and occupations, via the contraction principle. That provides an alternative to the existing approaches.

As a new and crucial quantity, unseen in close-to-equilibrium considerations, enters the traffic, measuring the time-symmetric dynamical activity in the system. This observable naturally enters beyond the linear response theory, *e.g.*, in determining the ratchet current [17] and in the escape rate theory, [18]. The overall traffic yields the current potential, and its excess together with an excess in the entropy flow directly determine the joint occupation-current fluctuations, (11).

The time-symmetric and time-antisymmetric fluctuations mutually couple even for small fluctuations around the nonequilibrium state, (27). Their decoupling in leading order around equilibrium is a fundamental reason for the known stationary variational principles to be approximately valid.

For extended systems with a large number of degrees of freedom, phase transitions may become visible through singularities of the fluctuation functionals, [19]. It should indeed not escape the attention that the analysis from (11) to (17) requires some strict convexity arguments and uniqueness of solutions. That is certainly one of the most fascinating possibilities that can be discussed within our general framework.

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- [1] L. Onsager S. Machlup, *Phys. Rev.* **91**, 1505 (1953).
 - [2] Yu. V. Nazarov (Editor), *Quantum Noise in Mesoscopic Systems*, Springer, 2003.
 - [3] C. Flindt, T. Novotný, and A. P. Jauho, *Europhys. Lett.* **69**, 475-481 (2005).
 - [4] S. Pilgram, A. N. Jordan, E. V. Sukhorukov, and

- M. Büttiker, *Phys. Rev. Lett.* **90**, 206801 (2003).
- [5] L. Bertini, A. De Sole, D. Gabrielli, G. Jona-Lasinio, and C. Landim, *Phys. Rev. Lett.* **94**, 030601 (2005).
- [6] T. Bodineau and B. Derrida, *Phys. Rev. Lett.* **92**, 180601 (2004).
- [7] B. Derrida, cond-mat/0703762.
- [8] D. J. Evans, E. G. D. Cohen, and G. P. Morriss, *Phys. Rev. Lett.* **71**, 2401 (1993).
- [9] G. Gallavotti and E. G. D. Cohen, *Phys. Rev. Lett.* **74**, 2694 (1995); *J. Stat. Phys.* **80**, 931 (1995).
- [10] C. Maes and K. Netočný, *J. Math. Phys.* **48**, 053306 (2007); S. Bruers, C. Maes, and K. Netočný, cond-mat/0701035, to appear in *J. Stat. Phys.*
- [11] C. Maes, K. Netočný and B. Wynants, cond-mat/0708.0489.
- [12] T. Fujisawa, T. Hayashi, R. Tomita, and Y. Hirayama, *Science* **312**:1634 (2006).
- [13] L. Bertini, A. De Sole, D. Gabrielli, G. Jona-Lasinio, and C. Landim, *Phys. Rev. Lett.* **87**, 040601 (2001).
- [14] M. D. Donsker and S. R. Varadhan, *Comm. Pure Appl. Math.* **28**, 1–47 (1975).
- [15] J. L. Lebowitz and H. Spohn, *J. Stat. Phys.* **95**, 333 (1999).
- [16] C. Maes and K. Netočný, *J. Stat. Phys.* **110**, 269 (2003); C. Maes, Séminaire Poincaré **2**, 29 (2003), Eds. J. Dalibard, B. Duplantier, and V. Rivasseau, Birkhäuser (Basel).
- [17] W. De Roeck and C. Maes, cond-mat/0610369.
- [18] P. Hänggi, P. Talkner, and M. Borkovec, *Rev. Mod. Phys.* **62**, 251 (1990).
- [19] Y. Kafri, E. Levine, D. Mukamel, G. M. Schütz, and J. Torok, *Phys. Rev. Lett.* **89**, 035702 (2002).